Single Spin Transport Spectroscopy - Current Blockade and Spin Decay

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We present a theory of a single-electron transistor exchange-coupled to a localized spin. We show how to gain detailed quantitative knowledge about the attached spin such as spin size, exchange coupling strength, Landé g-factor, and spin decay time T_1 by utilizing a robust blockade phenomenon of DC magnetotransport with accompanying noise enhancement. Our studies are of particular relevance to spin-resolved scanning single-electron transistor microscopy, electronic transport through nanomagnets, and the effect of hyperfine interaction on transport electrons by surrounding nuclear spins.

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The selective detection and manipulation of single spins are of fundamental importance for the realization of qubits as basic building-blocks in quantum computers[1]. In a solid-state environment, spins experience relaxation and decoherence which limit the functionality of quantum information processing. Therefore it is necessary to shed light onto the time scales of those effects. In addition, properties such as the coupling between spins as well as to an external magnetic field are important. Recently, the electrical time-resolved read-out of an individual electron spin enabled the measurement of the relaxation time[2] T_1. Furthermore, Wabnig *et al.* have proposed obtaining the T_1 and T_2 times by high-frequency (GHz) noise measurements[3]. Such experiments would be very challenging and we suggest here an alternative route to access at least T_1 which avoids time-resolved or high-frequency setups.

In this letter we explore the nonlinear current and low-frequency noise of a single-electron transistor (SET) where the spin of the quantum dot (QD) electron is coupled via the exchange interaction to a second localized spin. We find that tunneling spectroscopy in a constant magnetic field reveals a robust current blockade region due to population trapping that allows for the determination of T_1 in a simple manner. The proposed setup could be used as a spin-resolved version of the scanning single-electron transistor (SSET)[4], as alternative to single-spin detection by magnetic resonance force microscopy (MRFM)[5].

There are two further situations for which our work is of relevance: Electronic transport through nanomagnets (e.g. QDs doped with Mn-ions[6]) with an attached spin of 5/2 and an anisotropic exchange coupling; and as a starting point for studying the effect of hyperfine interaction on transport electrons by surrounding nuclear spins in QDs (e.g. as shown in Refs.[7,8] for double-QDs).

Our model consists of a SET with constant lead couplings $\Gamma_{L,R}$ and a QD electron spin $\vec{S}_2$ which is isotropically exchange-coupled to an additional spin $\vec{S}_1$ with strength J as sketched in Fig. 1a. The corresponding Hamiltonian (for the exchange term see also Ref.[9]), essentially an Anderson Model with exchange-coupled spin $\vec{S}_1$, reads $H = H_{\rm QD} + H_{\rm leads} + H_{\rm T}$, $H_{\rm leads} = \sum_{k\sigma\alpha=L,R} \varepsilon_{k\alpha} c^\dagger_{k\alpha\sigma} c_{k\alpha\sigma}$,

$$H_{\rm QD} = \sum_{i=1,2} \left(\varepsilon_i n_i + \mu g_i \vec{B}\cdot\vec{S}_i\right) + J\vec{S}_1\cdot\vec{S}_2, \qquad (1)$$

$$H_{\rm T} = \sum_{k\alpha\sigma_2} \left(t_{k\alpha\sigma_2} c^\dagger_{k\alpha\sigma_2} d_{\sigma_2} + {\rm h.c.}\right),$$

with ε_i the single-particle energy of the QD ($i = 2$) and the attached spin level ($i = 1$), $n_i = \sum_{\sigma_i} d^\dagger_{\sigma_i} d_{\sigma_i}$. Throughout this work the SET operates in the Coulomb blockade regime, i.e., only a single electron can enter the SET. This restricts the dimension of the accessible Hilbert space. The attached state with spin $\vec{S}_1$ is al-

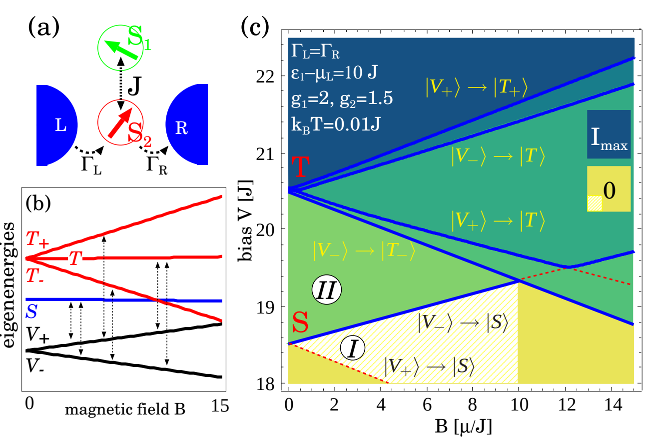

FIG. 1: (color online). (a) Sketch of the single-electron transistor (SET) with lead couplings $\Gamma_{L,R}$ and electronic level with spin $\vec{S}_2$ and attached spin $\vec{S}_1$ via exchange coupling J. (b) Eigenenergies of $H_{\rm QD}$ (singlet S, triplets $T, T_\pm$, unoccupied SET-level $V_\pm$) *vs.* magnetic field B for $g_{1,2} > 0$ (see Tab. I). Arrows indicate allowed transitions due to single-electron tunneling. (c) Steady-state SET current *vs.* symmetric bias voltage V and magnetic field B; Solid lines: current steps. Dotted lines: blocked transitions; Shaded region I: current blockade, bunched electron transfer (see also Fig. 2a). Region II: see text.

$\vert V_+\rangle \equiv \vert\uparrow 0\rangle$	$\varepsilon_1 + \varepsilon_z^{(1)}/2$
$\vert V_-\rangle \equiv \vert\downarrow 0\rangle$	$\varepsilon_1 - \varepsilon_z^{(1)}/2$
$\vert T_+\rangle \equiv \vert\uparrow\uparrow\rangle$	$\varepsilon + \varepsilon_z/2 + 1/4$
$\vert T_-\rangle \equiv \vert\downarrow\downarrow\rangle$	$\varepsilon - \varepsilon_z/2 + 1/4$
$\vert T\rangle \equiv \frac{1}{c_1}\left\{ c_3\vert\uparrow\downarrow\rangle + \vert\downarrow\uparrow\rangle \right\}$	$\varepsilon - 1/4 + c/2$
$\vert S\rangle \equiv \frac{1}{c_2}\left\{ c_4\vert\uparrow\downarrow\rangle + \vert\downarrow\uparrow\rangle \right\}$	$\varepsilon - 1/4 - c/2$

TABLE I: Eigenenergies and -states of H_{QD} (1) for $\vec{B} = B\vec{e}_z$, $\varepsilon_z^{(i)} \equiv \mu g_i B/J$, $\varepsilon \equiv \varepsilon_1 + \varepsilon_2$, $\varepsilon_z \equiv \varepsilon_z^{(1)} + \varepsilon_z^{(2)}$, $\Delta\varepsilon_z \equiv \varepsilon_z^{(1)} - \varepsilon_z^{(2)}$, $c_{1/2} \equiv \sqrt{1 + c_{3/4}^2}$, $c_{3/4} \equiv \Delta\varepsilon_z \pm c$, $c \equiv \sqrt{4\Delta\varepsilon_z^2 + 1}$. (All energies are scaled by the exchange coupling strength J throughout the paper. Realistic values for J can be estimated, e.g., by the dipole-dipole interaction between two electron spins[10] $J \approx (r_0/r)^3\, 0.1\mu\mathrm{eV}$ with distance r and $r_0 = 1\mathrm{nm}$.)

ways occupied with one electron. The Zeeman-energies $\mu g_i \vec{B}\cdot\vec{S}_i$ contain the Landé factors g_i and magnetic field $\vec{B}$. J denotes the exchange coupling strength. The operators $d_i^\dagger/d_i$ $(i = 1, 2)$ are the creation/annihilation operators in the attached spin/SET, and $c_{k\alpha\sigma}^\dagger/c_{k\alpha\sigma}$ are the creation/annihilation operators in lead $\alpha = L, R$ for momentum k and spin σ.

For the sake of simplicity and clarity we consider a spin $S_1 = 1/2$ throughout this work. The corresponding eigenenergies and -states of H_{QD} are presented in Tab. I, with the magnetic field-dependent spectrum shown in Fig. 1b. Note that the coefficients c_i $(i = 1\ldots 4)$ depend on the magnetic field. The Coulomb interaction between the QD electron and the side-electron simply produces an overall-shift of the spectrum and is therefore not considered further.

We treat the coupling to the contacts in second-order perturbation theory (sequential tunneling approximation). The standard Born-Markov-Secular approximation[11,12] for $B > 0$ (non-degenerate spectrum) leads to rate equations in the energy-eigenbasis. They can be cast into the compact form $\dot{\rho} = \mathcal{L}\rho$ with $\rho = \left(\rho_{(\uparrow 0)}, \rho_{(\downarrow 0)}, \rho_{T_+}, \rho_{T_-}, \rho_T, \rho_S\right)^T$ and $\mathcal{L} \equiv \mathcal{L}_L + \mathcal{L}_R$. The Liouvillians (6×6-matrices) $\mathcal{L}_{L/R}$ contain the couplings $\Gamma_{L/R}^\sigma = 2\pi\sum_k |t_{kL/R\sigma}|^2\delta(\varepsilon - \varepsilon_{kL/R\sigma})$, the Fermi functions $f_{L/R}(\epsilon_i)$ for the occupation of the L/R-contacts with the excitation energy ϵ_i and respective bias voltage $\pm V/2$, and the Clebsch-Gordan coefficients c_3/c_1, $1/c_1$, c_4/c_2, $1/c_2$ (see Tab. I). Counting fields $e^{\pm i\chi}$ are included in the right lead Liouvillian $\mathcal{L}_R = \mathcal{L}_R(\chi)$[13]. The cumulant generating function is given by $S(\chi, t) = \ln\left\{\mathrm{Tr}[\exp(\mathcal{L}(\chi)t)\bar{\rho}]\right\}$ with steady-state density matrix $\bar{\rho}$ given by $\mathcal{L}(0)\bar{\rho} = 0$. The cumulants are obtained via $C_k(t) = (-i)^k \partial_\chi^k S(\chi, t)\big|_{(\chi=0)}$. The steady-state current is $\langle I\rangle = e\dot{C}_1$ and the zero-frequency Fano factor (DC noise) is defined as $F \equiv \dot{C}_2/\dot{C}_1$. The Fano factor indicates sub(super)-Poissonian electron transfer if it is smaller(larger) than unity.

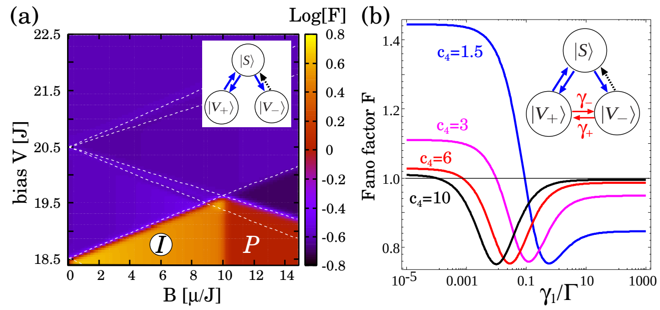

FIG. 2: (color online). (a) Fano factor *vs.* symmetric bias voltage V and magnetic field B, parameters as in Fig. 1c; region P: Poissonian charge transfer; Inset: configuration space with single-electron tunneling transitions for region I in Fig. 1c, Transition $|V_-\rangle \to |S\rangle$ is exponentially suppressed. (b) Fano factor *vs.* spin decay rate γ_1 for symmetric coupling $\Gamma = \frac{1}{2}\Gamma_{L/R}$ and various coefficients $c_4 \propto B$ in region I (see Fig. 2a); Inset: Configuration space with indicated spin flip rates.

The spectrum of H_{QD} (Tab. I) itself is not directly accessible by transport spectroscopy. Rather, by applying a bias voltage V to the SET, the excitation spectrum is probed. The six transitions with nonvanishing Clebsch-Gordan coefficients are indicated in Fig. 1b. Fig. 1c shows that the current *vs.* magnetic field B and bias voltage V is a direct map of those transitions. The six steps correspond to excitations in the eigenspectrum (assuming the temperature is low, otherwise the transitions would be smeared out). Between steps the current is nearly constant for fixed, but changes with variable magnetic field due to the dependence of the Clebsch-Gordan coefficients. At zero-magnetic field, two current steps occur corresponding to tunneling transitions from the unoccupied states to the singlet- and degenerate triplet states. The bias difference of the steps is $2J$. From the positions of the current steps one easily obtains the g-factors of both spins by fitting the excitation energies calculated by the differences of eigenenergies in Tab. I. We point out that the specific dependence of the excitation spectrum on the magnetic field can appear very different when one of the g-factors is negative (as widely occurs in confined electron systems) e.g. for $g_1 < 0$ negative differential conductance appears for certain transitions.

Remarkably, certain transitions are completely missing (dotted lines in Fig. 1c) even though the corresponding Clebsch-Gordan coefficient is not zero. This is to be contrasted with spin blockade for which the relevant coefficients disappear[14]. This blockade here is caused by the topology of the configuration space. Let us focus on the lowest lying transition in Fig. 1c where the configuration space consists of three states: $|V_+\rangle$, $|V_-\rangle$, and $|S\rangle$ in the inset of Fig. 2a. Since the transition energy for $|V_-\rangle \to |S\rangle$ is too large to be supplied by the lead electrons in that bias range, the rate for that process is exponentially suppressed. Consequently, the system gets trapped in the state $|V_-\rangle$ once it reaches

there, the spin S_1 is completely polarized and the current is blocked. Such population trapping effect can be expected in any system with a configuration space with one or more singly-connected states. We emphasize that the current blockade persists for an arbitrary choice of J, $g_{1,2}$, S_1, and even for ferromagnetic leads $\Gamma^{\uparrow}_{L,R} \neq \Gamma^{\downarrow}_{L,R}$. However, the blockade will be removed with increasing temperature T since the transition $|V_-\rangle \to |S\rangle$ becomes more likely (compare inset of Fig. 2a). In regions I and II the dynamics can be described by an effective three-state model assuming low temperatures such that $f_R(\epsilon_{s1}) = f_R(\epsilon_{s2}) = 0$ and excitations involving triplet states are negligible:

$$\begin{aligned}
\dot\rho_{|\downarrow 0\rangle} &= \{-\Gamma_L f_L(\epsilon_{s1})\rho_{|\downarrow 0\rangle} + [\Gamma_L f_L^-(\epsilon_{s1}) + \Gamma_R]\rho_{|S\rangle}\}c_2^{-2},\\
\dot\rho_{|\uparrow 0\rangle} &= \{-\Gamma_L f_L(\epsilon_{s2})\rho_{|\uparrow 0\rangle} + [\Gamma_L f_L^-(\epsilon_{s2}) + \Gamma_R]\rho_{|S\rangle}\}\frac{c_4^2}{c_2^2},\\
\dot\rho_{|S\rangle} &= \{\Gamma_L f_L(\epsilon_{s1})\rho_{|\downarrow 0\rangle} + c_4^2\Gamma_L f_L(\epsilon_{s2})\rho_{|\uparrow 0\rangle}\\
&\quad - [\Gamma_L[f_L^-(\epsilon_{s1}) + c_4^2 f_L^-(\epsilon_{s2})] + \Gamma_R c_2^2]\rho_{|S\rangle}\}c_2^{-2} \qquad (2)
\end{aligned}$$

with $\epsilon_{s1/s2} \equiv \varepsilon_2 \pm \varepsilon_z^{(1)}/2 - 1/4 - c/2$ and $f_L^-(\cdot) \equiv 1 - f_L(\cdot)$.

The Fano factor in region I is $F_I = 1 + 2c_4^2\Gamma_R/(\Gamma_L + \Gamma_R)$ and clearly indicates super-Poissonian electron transfer since $c_4 > 1$ for $B > 0$ (lower left corner of Fig. 2a). A very similar expression has been derived in Ref.[15] with the interaction parameter Φ replacing c_4. The underlying mechanism is related to thermally-activated bunching of tunneling events[16]: as long as the system is in state $|V_-\rangle$ no tunneling occurs. However, with an exponentially small probability, thermally-excited lead electrons can enter the state $|S\rangle$ and a small bunch of tunneling transitions $|V_+\rangle \leftrightarrow |S\rangle$ may take place. Outside of region I the noise is Poissonian (P) or sub-Poissonian as can be seen in Fig. 2a for symmetric lead couplings $\Gamma_L = \Gamma_R$. For asymmetric couplings, it is also possible to observe $F > 1$ in other regions, e.g., in region II when the condition $\Gamma_L/\Gamma_R > (c_4 - 1)^2/\,2c_4$ is fulfilled.

We can exploit the current blockade mechanism in order to measure the spin relaxation rate $\gamma_1 = 1/T_1$ of S_1. In the inset of Fig. 2b we show that the formerly singly-connected state $|V_-\rangle$ is additionally linked with state $|V_+\rangle$ via spin flip transitions with rates $\gamma_+ = \gamma_1 f(\varepsilon_z^{(1)})$, $\gamma_- = \gamma_1[1 - f(\varepsilon_z^{(1)})]$. Such a mono-exponential spin decay can occur, e.g., due to spin-orbit coupling[17]. The steady-state current is then directly related to the spin-relaxation rate $1/T_1$ and it can be simply measured by the ratio of plateau currents in regions I and II at the same magnetic field (assuming $\gamma_1 \ll \Gamma$ so that $\langle I\rangle_{II}$ does not depend on γ_1):

$$\gamma_1^{-1} \;=\; A_1\left[A_2\frac{\langle I\rangle_{II}}{\langle I\rangle_I} - A_3\right] \qquad (3)$$

with $A_1 \equiv c_2^2/(\Gamma\Gamma_L c_4^2)$, $A_2 \equiv [1 - f(\varepsilon_z^{(1)})](\Gamma_L + 2\Gamma_R)$, $A_3 \equiv (c_4^2\Gamma + \Gamma_L[1 - f(\varepsilon_z^{(1)})] + \Gamma_R)$, and $\Gamma \equiv \Gamma_L + \Gamma_R$.

The Fano factor *vs.* γ_1 in Fig. 2b shows that by increasing the spin relaxation the charge transfer turns from super- to sub-Poissonian. For large magnetic fields the Fano factor approaches unity: for small γ_1 from the super-Poissonian side and for large γ_1 from the sub-Poissonian side. Hence, the zero-frequency noise provides a sensitive quantitive indicator of the spin decay.

In region I the exponential suppression of the current can also be lifted by algebraic cotunneling contributions[18]. To ensure that the effect of spin relaxation on the SET transport is not obscured by higher-order tunneling processes, the rates $\Gamma_{L/R}$ have to be small with respect to the temperature.

The discussed effects are robust against background charge fluctuations. We have checked that by considering fluctuating energy levels $\varepsilon_i(t) = \varepsilon_i + 1/\sqrt{\tau_i}\,\xi_i(t)$ $(i = 1,2)$ with $\langle\xi_i(t)\rangle = 0$ and $\langle\xi_i(t)\xi_j(t')\rangle = \delta_{ij}\delta(t - t')$. Second-order perturbation theory in the fluctuation strength leads to the Lindblad form[11] $\mathcal{L}_{\mathrm{cf}}\rho = \sum_i \tau_i^{-1}[n_i\rho n_i - \frac{1}{2}\{n_i^2, \rho\}]$, which leaves the relevant subspace invariant.

In summary, we have studied a Coulomb-blockaded SET with exchange-coupled 1/2-spin in a constant magnetic field. The excitation spectrum (current *vs.* magnetic field and bias voltage) shows rich structure enabling the measurement of the exchange-coupling strength J and the Landé g-factors. Due to classical population trapping the lowest-lying transition is blocked independent of J, the g-factors and the spin size - the current is exponentially suppressed and the electron transfer is super-Poissonian. As we demonstrate, this robust phenomenon allows the direct measurement of the decay rate $1/T_1$ of the probed spin by DC current and noise measurement. Our method can be readily used to implement larger S_1 encountered in nanomagnets or to describe the nuclear spin environment of a QD.

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